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THE DISTRIBUTION OF RADIOACTIVITY FROM RAIN

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Cincinnati, Ohio

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THE DISTRIBUTION OF RADIOACTIVITY FROM RAIN*

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Fallout measurements, largely as precipitation, have been made on a continuous basis at Cincinnati, Ohio, since March 1953. The studies of 1953 (1) and 1954 (2) were implemented in October 1953 by weekly or daily tests of the atmospheric particulate activity and by the operation of an experimental cistern near the site of the rain collector. Correlation is now possible between air particle concentration and rainout (washout by precipitation) and distribution of activity between the separated solids and the supernatant liquid in the cistern liquid can be determined. This report includes data relating to activity levels in air particulates, rainfall, and certain Ohio rivers.

METHODS

The methods of sample collection have been described (2). The precipitation values reported in Figures 1 and 2 are the means of values reported for six stations and were obtained from U.S. Weather Bureau

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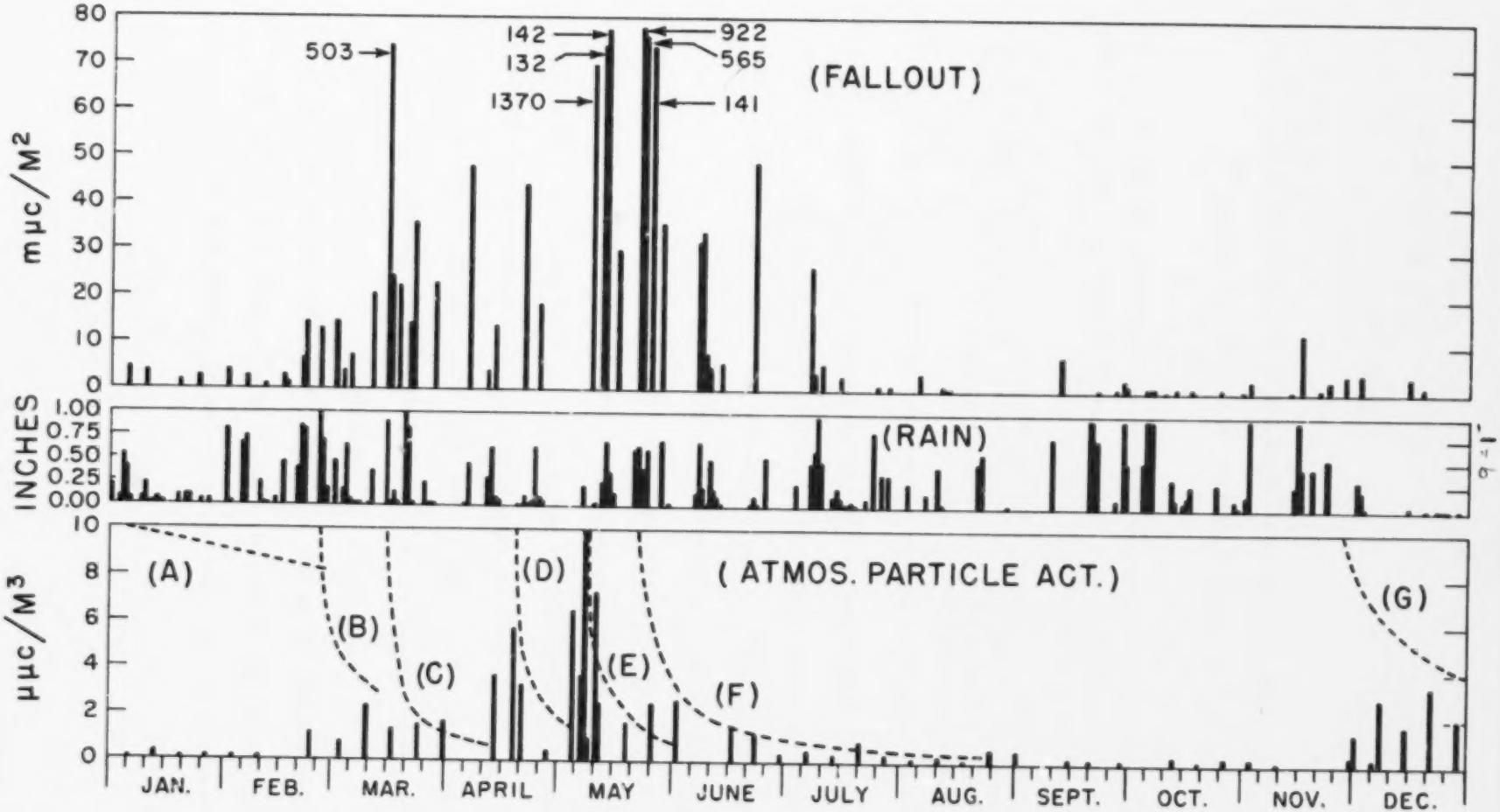
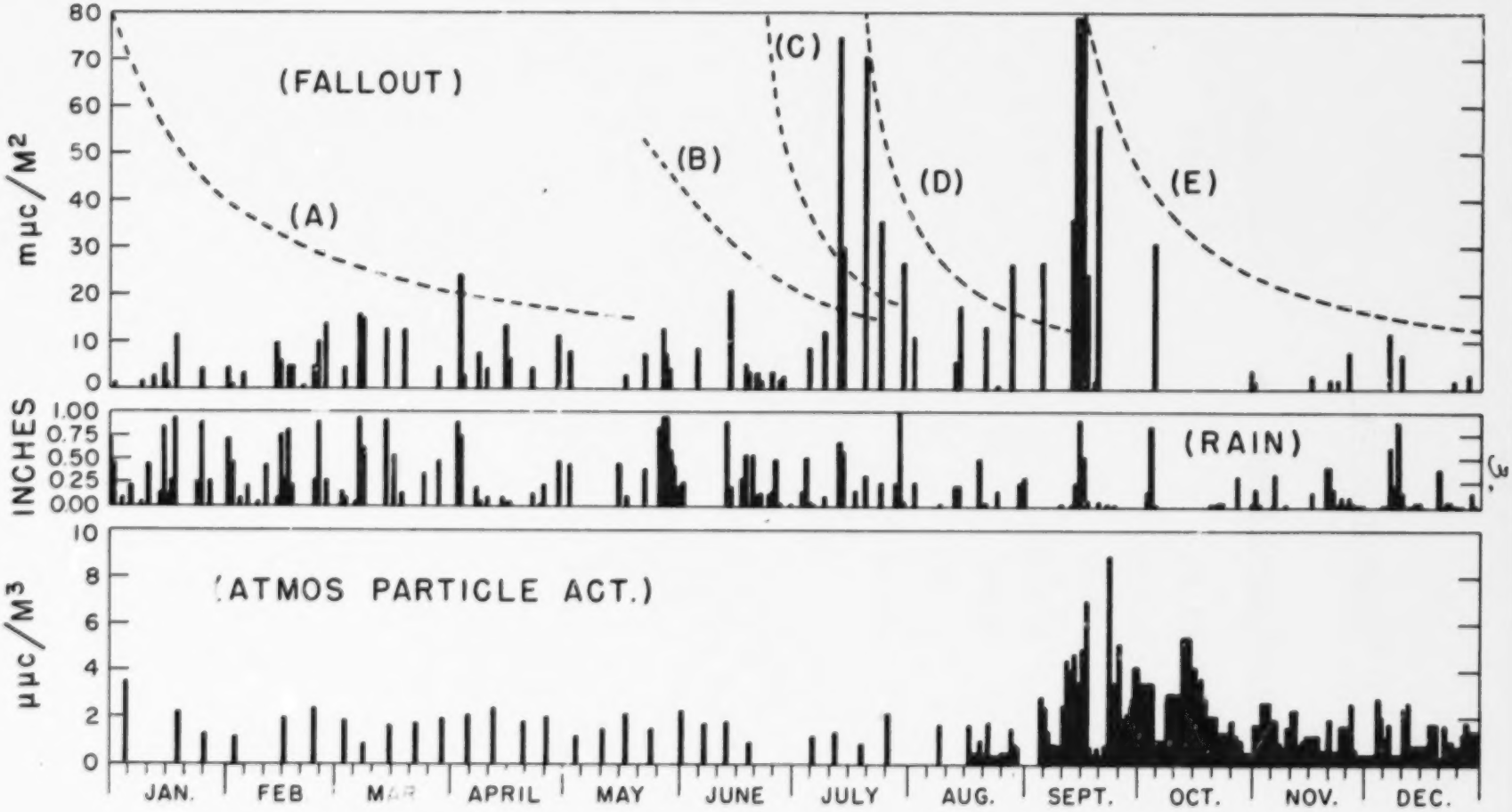


RADIATION LEVELS, AIR AND RAINFALL 1955

FIGURE 1

LEGEND FOR FIGURE 1

	Date Bomb Debris Formed	Decay Slope
a.	6/4/54	1.0
b.	2/18/55	1.2
c.	3/12/55	1.0
d.	4/15/55	1.2
e.	5/5/55	1.2
f.	5/15/55	1.2
g.	11/5/55	1.0



RADIATION LEVELS, AIR AND RAINFALL 1956

FIGURE 2

LEGEND FOR FIGURE 2

	Date Bomb Debris Formed	Decay Slope
a.	12/1/55	1.0
b.	5/3/56	1.0
c.	6/13/56	1.0
d.	7/10/56	1.0
e.	8/27/56	1.0

climatological data. To compensate for differences in hours of collection some adjustment in these values was made.

Rainfall was collected in a pail-type sampler. Ailquot samples were filtered through membrane filters and the filtrates evaporated. The gross beta-gamma activity of the filter and dissolved solids was measured, and the results reported on an area basis. Repeat measurements on many samples were made to establish the decay rates of suspended and dissolved activities. When combined the suspended and dissolved activity usually exhibited an exponential negative decay rate of from 1.0 to 1.2 for known dates of bomb tests. For tests of unknown origin dates were selected to conform in general to this rate of decay.

The air particulate activity of samples from January 1, 1955, to August 1956 was determined from swatch sub-samples representing roughly 80 cubic meters of air filtered through glass fiber filters over a 24-hour period, from the Lincoln Park area, Cincinnati (3). Continuous air sampling was initiated August 1, 1956. Air activity was obtained daily (except week-ends) by filtering 20 to 30 cubic meters of air through a membrane (cellulose acetate) filter. Weekend samples were collected after one to three. The membrane filter sample was placed in an aluminum dish, saturated with 95 per cent alcohol, ignited, flash flamed with a Meeker burner, and counted for two 8-min. periods in an internal proportional counter. The beta counting was usually postponed for 2 days after collection to prevent the interference of short-lived naturally-occurring alpha and beta activity (4).

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RESULTS

Precipitation, Air Particulate and Fallout Activity

The results of precipitation in inches, air particulate activity in micromicrocuries per cubic meter ($\mu\mu c/M^3$), and the fallout activity in millimicrocuries ($m\mu c$) per square meter have been summarized in Figure 1 (1955 data) and 2 (1956 data). From the decay of rain samples the age of the activity was estimated and is shown for most of the samples in the legends of Figures 1 and 2. The rate of decay is plotted as broken lines in Figures 1 and 2.

Considering the particulate activity in air from stations in the Midwest (Cincinnati, Louisville, St. Louis, Chicago, Minneapolis, and Detroit) the average background level of approximately 0.05 $\mu\mu c/M^3$ in the fall of 1953 has increased to an arithmetic average level of 0.6 $\mu\mu c/M^3$ in 1954, 1.55 $\mu\mu c/M^3$ in 1955, and 2.26 $\mu\mu c/M^3$ in 1956. During continental tests in 1955, the activity in Cincinnati air (Figure 1) fluctuated from a high of 10 $\mu\mu c/M^3$ of rapidly decaying radioactivity to a low of 0.08 $\mu\mu c/M^3$. Due primarily to the more distant bomb test areas and the consequent dispersion of activity and decay of short-lived isotopes there was less fluctuation in radioactivity levels (8.9 to 0.58 $\mu\mu c/M^3$) during the Pacific and foreign tests of 1956. However, a higher sustaining level of air radioactivity was noted for the entire year.

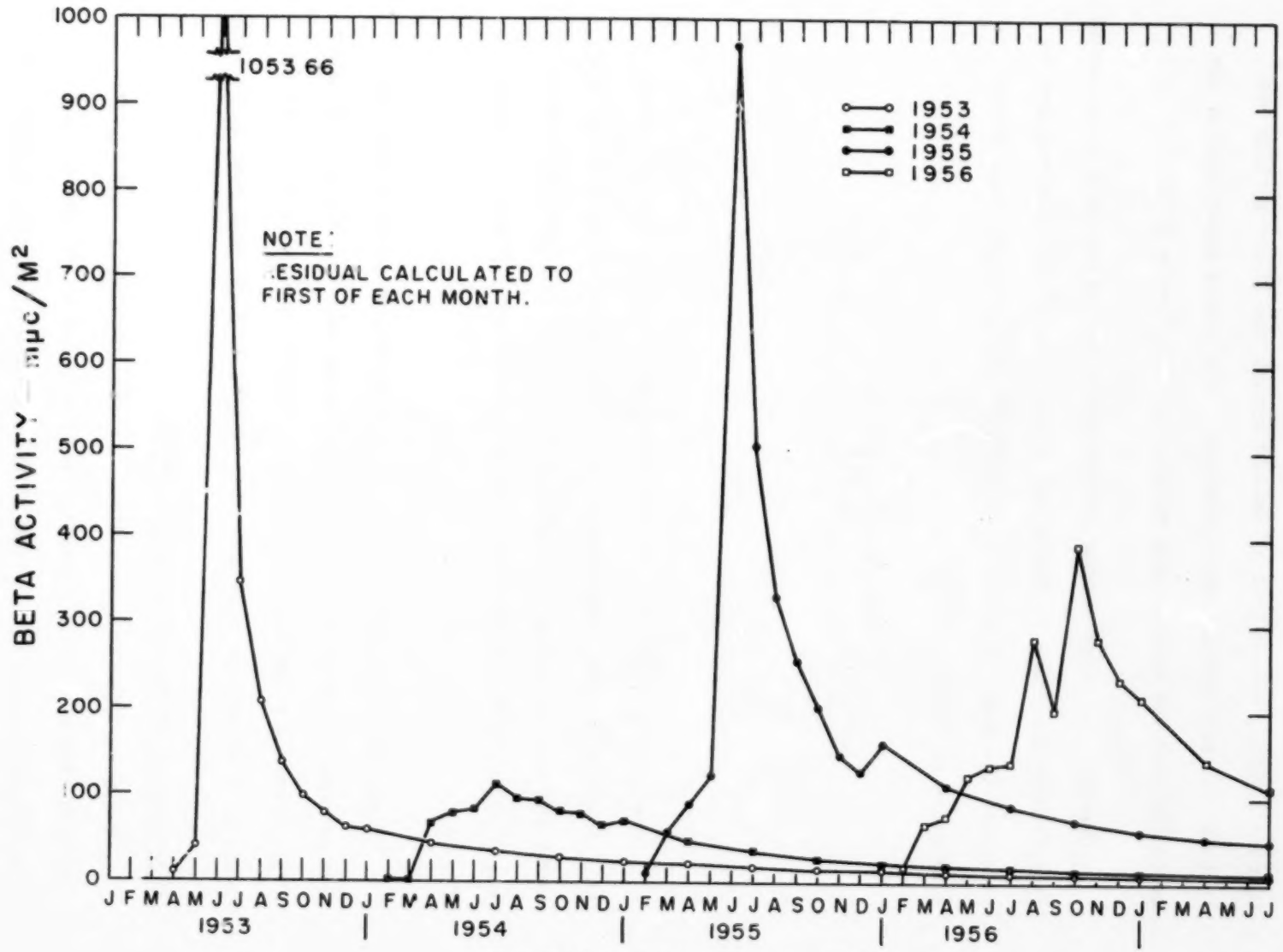
Fallout from 15 rain collections made between March 15 and June 25, 1955, showed activity levels above 30 $m\mu c/M^2$, with the level in seven of these varying from 141 to 1370 $m\mu c/M^2$ (Figure 1). Most of this activity was short lived as shown in the decay curves. Relatively little

radioactivity was observed in samples collected during the first two and last six months of 1955, but the activity collected was of longer half-life. Although the air was relatively active in December, precipitation and consequently the fallout was insignificant.

In 1956 (Figure 2) the radioactive fallout in excess of 10 $m\mu c/M^2$ was common throughout the year. During this time air activity was moderately low (less than 3 $\mu\mu c/M^3$) except in September and October. The decay curves show that most of the fallout activity in 1956 was longer-lived than that collected in 1955.

A somewhat better appraisal of the amount of long-lived radioactive fallout may be obtained by considering the cumulative residual deposition. At Cincinnati, and presumably at all non-arid areas several hundred miles removed from the weapons test site (5), increased radioactive fallout appears to be associated with periods of rainfall and snowfall.

The radioactive fallout, identified quite precisely as to age and rate of decay for the more radioactive samples and less accurately for the low level radioactive samples, has been extrapolated to the first of each succeeding month. The cumulative residual activity in millimicrocuries per square meter ($m\mu c/M^2$) of earth's surface, assuming no loss, is plotted as shown in Figure 3 for each of the four years of record from April 1953 to July 1, 1957. In each year there is a peak of activity during or immediately following test operations. High peaks of 1054 $m\mu c/M^2$ on June 1, 1953 and 970 $m\mu c/M^2$ on June 1, 1955, are shown for the two years of continental tests (1953 and 1955), whereas longer lived peaks of 110 and 390 $m\mu c/M^2$ were observed on July 1, 1954, and



RESIDUAL FALLOUT

FIGURE 3

October 1, 1956, respectively. Fallout during the latter periods was from Pacific and foreign test operations. The yearly deposition of observed fallout was extrapolated to one-year old fission products.

One-year old mixed fission products contain 3.9 per cent strontium-90 (yttrium-90) according to Hunter and Ballou (6). This value was used to calculate the combined activity of Sr^{90}-Y^{90} deposited for each of the four years as follows:

Year of Deposition	One-year-old activity $\mu\mu c/M^2$	Sr^{90}-Y^{90} $\mu\mu c/M^2$
1953	31.3	1.22
1954	37.5	1.46
1955	63.2	2.46
1956	150.4	5.87
Total '53-'56	282.5	11.0

Precipitation for each of the four years amounted to 22.9, 36.3, 44.7 and 41.8 inches, respectively or a total of 145.7 inches. On this basis the average activity of precipitation over the four-year period was 76.4 $\mu\mu c/l$ of one-year-old fission product and 3.0 $\mu\mu c/l$ of Sr-90 (Y-90).

These levels in water appear insignificant, compared to the maximum permissible level of Sr^{90}-Y^{90} in drinking water of 800 $\mu\mu c$ per liter (7). However, it may be significant to observe the increase in the deposition from year to year which has roughly doubled each year since 1954.

Distribution of Fallout in Cistern Water

Early in 1954, an experimental cistern, consisting of a hopper-bottomed tank of 2.62 M^3 capacity, was installed to collect precipitation from a portion of the roof of an abandoned home. The cistern was

in operation from February 10, 1954, to March 7, 1956, and from July 5 to September 25, 1956. During this period, settle cistern water was withdrawn and tested at frequent intervals while the settled sludge was removed and tested on fifteen occasions. The more significant data from cistern operation are presented in Table 1. The total activity found in the cistern water, sludge, and sludge supernatant are indicated in Column 7. Sludge supernatant consisted of a known quantity of cistern water (up to 50 liters) used to wash the sludge from the cistern bottom. This water was then settled for 10 to 15 minutes and the supernatant removed from the sludge by decantation. The sludge contained loose shingle stone which had been washed into the cistern. It was separated from the sludge and samples of the supernatant, sludge and roof stone were tested for beta radioactivity. The sludge and supernatant solids found in the roof drainage varied from 9 to 71 with an average of 30 ppm of dry solids of which 30 per cent were volatile.

During the sixteen month period of cistern operation, a total of 50.4 uc of beta activity (Column 6) from bomb debris 10 to 300 days old was estimated to be in the cistern rain. Of this total, 36.72 μc or 73 per cent was recovered from the cistern. The remaining 27 per cent represents activity fixed to the asphalt roof shingles and errors in measurement. It is interesting to note that estimations based on the activity found on washed shingle stone separated from the cistern sludge accounted for much more than 27 per cent unaccounted for activity. However, since the stones tested were bathed in radioactive rain for days or weeks they may have adsorbed more activity than would shingle stones during roof runoff.

TABLE 1

The Distribution of Fallout in Cistern Water

Series No.	Date of Accounting	Precip. Inches	Ext. Date*	Age of Bomb Debris Days	Total Precip. Inches	Gross Beta Radioactivity: Recovered in Cistern μc	In Rain μc	Dissolved in Cist. Water %	Suspended in Cist. Water %	In Cistern Water: Dissolved μμc/L	Total μμc/L
1	3/4/55	19	3/8	19-300	6.34	2.86	1.40	14	77	69	445
2	3/16	5	3/19	7-30	1.37	10.00	9.71	53	22	4160	5890
3	3/21	4	3/22	10	1.96	1.02	0.95	30	17	163	256
4	5/5	16	5/11	20-55	2.79	1.46	2.15	46	13	262	337
5	5/17	6	5/26	15	1.59	3.48	7.16	28	12	660	950
6	6/21	14	6/29	37	4.62	8.02	8.60	38	33	720	1340
7	7/14	11	7/20	60	3.68	1.56	2.07	56	1	259	263
8	8/24	16	8/26	101	3.07	0.36	0.28	26	7	34	43
9	9/28	6	10/4	136	3.50	0.15	0.34	49	7	22	25
10	10/25	11	11/10	163	6.01	0.17	0.20	23	8	7	10
11	11/18	6	11/30	187	2.93	0.11	0.53	39	5	24	23
12	2/9/56	36	2/9	64-270	9.32	1.16	1.14	56	34	76	123
13	3/7	11	3/7	36-98	5.08	1.65	1.83	37	27	157	269
14	7/27	8	7/27	17-44	2.08	4.14	4.66	19	23	410	908
15	9/25	24	9/25	15-77	5.58	7.81	9.40	55	22	762	1070
1-15	Total	193			59.92	43.94	50.40	40**	25**	322**	524**

* All measurements in a series extrapolated for decay to this date.

** Obtained from activity (total or dissolved) ÷ total rainfall.

The soluble activity in the cistern water varied from 14 to 55 per cent or a weighted average of 48 per cent of the total. There seems to be no clear cut relationship between the percentage of dissolved activity and the age of bomb debris. This is contrary to the observation made in 1953 (1) that the percentage of dissolved activity in the rain was less for activity of short half-life. Other factors, such as the abundance of non-radioactive air pollutants, the time of exposure of radioactive particles, and possibly the inflow of radioactive contaminants of different chemical characteristics from the stratosphere to the rainout zone, probably play a role in forming the soluble fraction.

The concentration of dissolved activity in cistern water varied from 7 to 4160 or an average of 332 μμc/l. Lower values (less than 76 μμc/l) were observed during periods when the bulk of the activity was estimated to be older than 100 days, whereas for activity 7 to 98 days in age, the dissolved activity was 157 μμc/l or more. The percentage of activity associated with the suspended solids ranged from 1 to 77 per cent, with an average of about 25 per cent.

In summary the cistern study reveals that slightly more than one-half of the activity found in the cistern water was soluble and that up to a third of the activity would be associated with the suspended solids. Over two thirds of the insoluble activity settles as a sludge (1.5 kg dry weight) having an activity of 10 μc/kg (dry weight). Septicity resulted in further solution of activity, but diffusion into the cistern water was minimal when the sludge was undisturbed.

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The Distribution of Fallout in Surface Waters

Nine grab and seven composite (7 to 10 day) samples from 13 stations on the Ohio River from Pittsburgh, Pennsylvania, to Louisville, Kentucky, were collected between December 27, 1955, to April 20, 1956, in a cooperative study with the Ohio River Valley Water Sanitation Commission. The maximum beta activity found was 100 μμc/l as compared to an arithmetic average value of 27.5 μμc/l of which 15 μμc/l was in solution.

Through the courtesy of F. H. Waring, Chief Sanitary Engineer, Division of Sanitary Engineering, Ohio State Board of Health, the results of additional samples collected from surface streams tributary to the Ohio River are included in Table 2 and Figure 4. The maximum average activity levels of the surface waters of the Little Miami, the Great Miami, and Scioto Rivers are compared with the SEC Cincinnati rainout results for collections made between river samplings.

The stream water results show a maximum dissolved activity of 89 μμc/l compared to a maximum total (suspended and dissolved) activity of 217 μμc/l for tests in 1956. The average total activity for each sampling date fluctuated from 25 to 130 μμc/l depending to some degree on the activity of the more recent rains. The yearly geometric average total and dissolved beta activity of stream water was 50 and 27 uuc/l respectively, as compared to 974 μμc/l total and 389 μμc/l dissolved activity for Cincinnati rain. The average removal by natural agents is therefore 95 and 93 per cent for total and dissolved activity, respectively. Actually, the removals of rain activity may be even greater inasmuch as some of these streams are known to receive some radioactive wastes from industrial, research and medical facilities.

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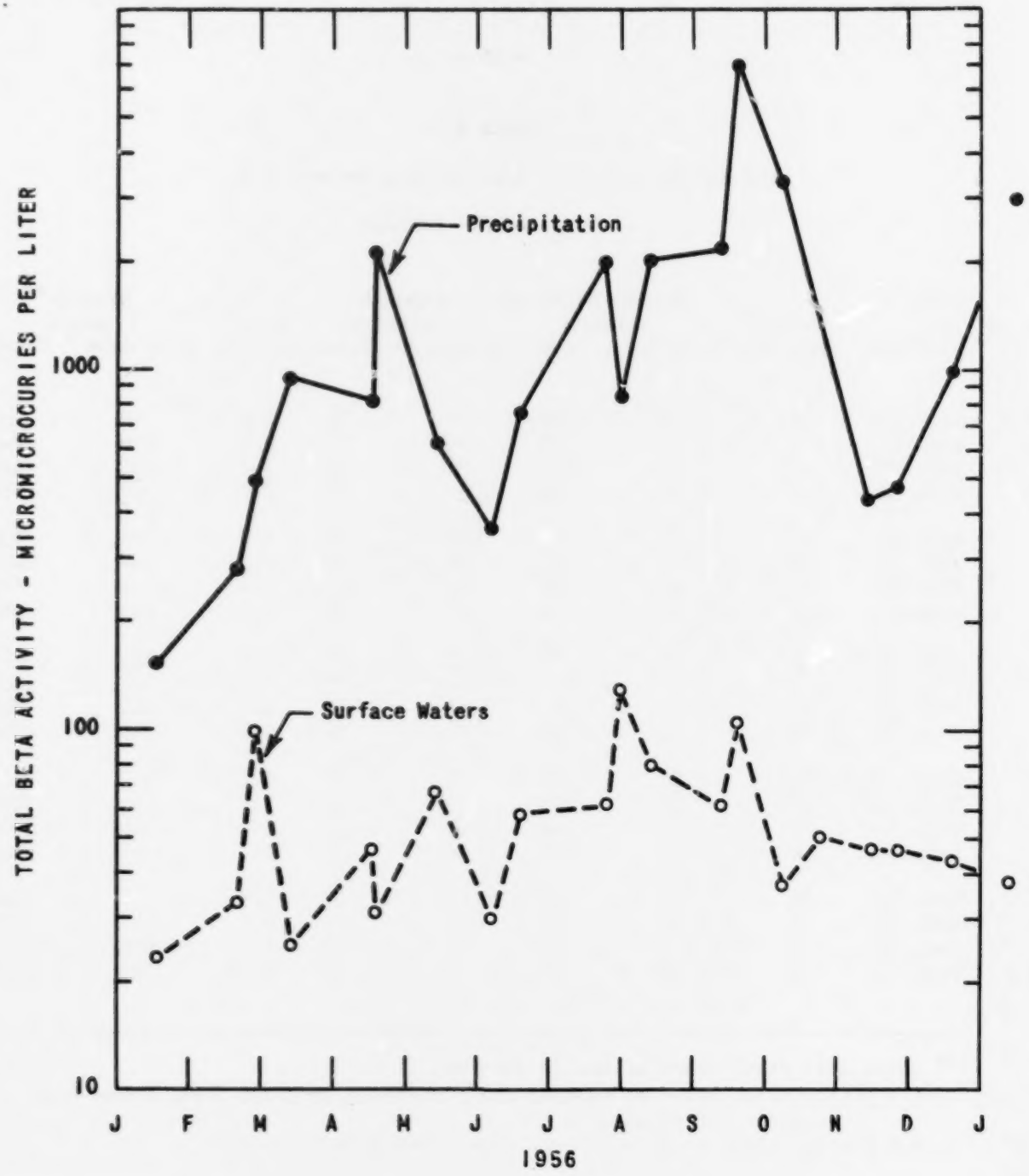


A COMPARISON OF CINCINNATI FALLOUT IN RAIN
WITH THE ACTIVITY OF OHIO STREAM WATER

Figure 4

TABLE 2

The Radioactivity of Ohio Surface Water - 1956

as Compared to Rainout

Date of Collection	Stream* Stations	Stream Water Activity-μμc/L Maximum Dissolved	Maximum Total	Average Dissolved	Average Total	Rain Inches	Rainout** μμc/L Dissolved	Rainout** μμc/L Total
Jan. 17	A	19	42	6	23	2.5	50	150
Feb. 20	A	—	43	—	33	6.8	160	280
Feb. 28	B	80	141	52	99	1.2	100	490
Mar. 13	A	—	38	—	25	2.0	360	950
Apr. 17	B	38	54	29	46	3.7	—	830
Apr. 24	A	25	40	16	31	0.2	—	2120
May 14	A	64	87	43	67	1.2	—	620
June 6	B	29	50	10	30	4.6	140	360
June 18	A	30	66	23	58	1.1	420	760
July 25	B	45	79	44	62	5.0	—	2000
July 31	A	80	217	64	130	1.3	510	840
Aug. 15	A	51	140	32	80	0.6	1310	2040
Sept. 11	A	84	116	35	62	1.2	1420	2180
Sept. 19	B	89	178	57	105	1.7	2260	7000
Oct. 8	B	27	44	22	37	1.1	1320	3300
Oct. 23	A	30	158	20	51	0	—	—
Nov. 14	B	36	46	31	47	0.5	317	440
Nov. 26	A	60	68	39	47	0.6	—	478
Dec. 18	A	34	69	23	44	1.09	624	992
Jan. 15	B	26	42	20	38	1.42	193	2941

* Note: Ohio State Board of Health data on,
A - Little Miami River at Newtown and 5 stations on Great Miami River from river mile 24.8 to 96.6
B - Four stations on Scioto River from river mile 15 to 140.

** Weighted average activity of rain at Cincinnati collected between dates of stream sampling.

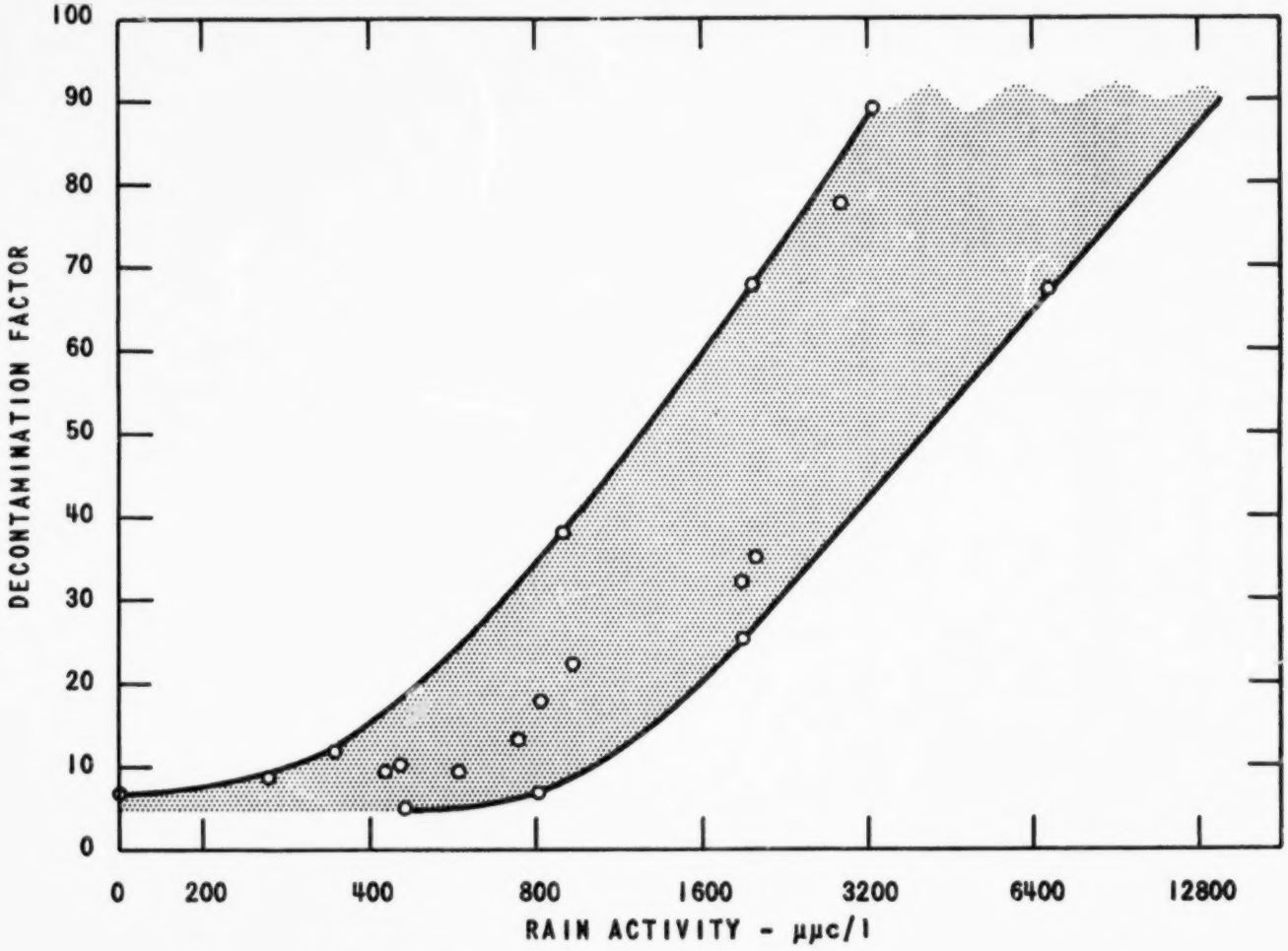
The plotted results (Figure 4) more clearly define the relationship of total rain activity (from Cincinnati fallout) with that found at stream stations up to several hundred miles distant. Obviously, Cincinnati rain and activity measurements may not be representative of the rainout at some of these stream stations. Nevertheless, general trends may be observed and the desirability of establishing more rain monitoring stations is apparent. The rain data indicate a sharp increase (150 to 2100 μμc/l) during the first of the year, then, following a drop in May and June, a second rise to levels of 2000 to 7000 μμc/l by September. During the last three months of the year, the rain activity decreased. The lower levels of rain activity usually correspond to longer-lived bomb debris and high levels correspond to young (rapidly decaying) fission products.

The ratio of total rain activity to total surface water activity was determined for each month and reported in terms of the decontamination factor (rainout or fallout activity divided by the surface water activity). These decontamination values were plotted against the rain activity as shown in Figure 5. It will be seen that removal of radioactivity by natural agents increases with increased activity of the rain. Comparable in many respects with other pollutants, the plotted values indicate the application of the law of diminishing returns, i.e., high decontamination factors in the order of 65 to 90 (98.5 to 98.9 per cent removal) for relatively young bomb debris and decontamination factors of 5 to 10 (80 to 90 per cent removal) for lower-level longer-lived activity.

The Cincinnati rainout and cistern studies have been combined with the Ohio State stream data for comparison of the distribution of radioactive rainout. The rainout (corrected for decay to the end of a cistern sampling period), the cistern water activity of series 11 to 15 (Table 1), and the activity of stream waters of appropriate date are summarized in Table 3. The table shows that subsidence in the cistern and adsorption by roof shingles results in an average loss of 38.5 per cent of the total and 20.6 per cent of the dissolved rain activity, whereas the low activity found in stream waters indicatesthat radioactive particles adsorbed on surfaces, such as vegetation and soil, and percolation and dilution play important roles in achieving a 92 and 91 per cent reduction of the total and dissolved beta activity of rain, respectively.

Discussion

Radioactive fallout has been under close surveillance since nuclear weapons were first tested. Reports by Libby (8, 9), and Eisenbud and Harley (10) are based on gummed film collections. Analysis of data by List (11) shows that a preponderance of the fallout obtained with gummed films occurs during precipitation. Our findings show that about one-half of fallout activity is dissolved in rain water. Similarly about one-half of the activity collected from dust collectors following Operation Hurricane, as reported by Russel et al (12) was found to be soluble in distilled water and dilute salt solutions. The fraction of the total activity which is dissolved or soluble depends on many factors such as the age and particle size of the bomb debris and the adsorptive capacity of local atmospheric dust with which it comes in contact before or during rainout.



THE DECONTAMINATION OF RAIN BY NATURAL AGENCIES

Figure 5

TABLE 3

Comparative Beta Activity of Rain, Cistern and Stream Waters

(Results in micromicrocuries per liter)

Series Dates	Dissolved Activity			Total Activity		
	Rain	Cistern	Streams	Rain	Cistern	Streams
11/18-2/9	54	76	6	130	123	23
2/9-3/7	152	157	52	390	269	33-99
7/5-7/27	1430	410	44	2420	908	62
7/27-9/25	800	762	32-64	1830	1070	62-130
Geometric means	311	247	28.4	689	424	54.8
% Removal compared to rainfall		20.6	91		38.5	92

Although significant radioactive dust-fall of the larger particles may occur in the vicinity of a nuclear test site the finer particles are removed from the air by rainout or snowout and, to a lesser degree, by the inpingement of particles on leaves, grasses and other solid surfaces by wind action. A comparison of the estimated $m\mu c/M^2$ of strontium fallout from rain collections at Cincinnati, Ohio, New York City, New York, (13), and Milford Haven, England (14) is as follows:

Year of Deposition	Cincinnati Ohio	New York City New York	Milford Haven England
1953	1.22	—	—
1954	1.46	2.88	1.15 a
1955	2.46	3.35	2.65
1956 (1st Half)	2.8	6.67	0.5 b
1956 (Last Half)	3.07	—	—

Note:
The Milford Haven results (HP/R 2017) are reported in units 1000 times higher but the concentration in rain water and a subsequent discussion of these results (HP/R 2056) verifies the correctness of the $m\mu c/M^2$ unit.

a-May through December
b-January through March

Agreement in fallout values is surprisingly good for the three stations in 1954 and 1955. Disagreement of values for the first five or six months of 1956 may be expected from the known variations in weather patterns with respect to the presence of radioactive air masses prior to more uniform distribution with time. This was clearly demonstrated in Cincinnati during December 1955 when air activity was relatively high but, since no precipitation occurred, the fallout was negligible.

In order to determine the variation in fallout or its distribution over grazing or farm land many more collectors will be necessary. Many rain collectors should be satisfactory, such as the 13.5 inch bucket used, the 8-inch pot Harley et al (13) used in New York or the 0.22 square meter polyethylene funnel and bottle used at Milford Haven (14). In continuous use each will collect true dust-fall and secondary aerosols (from previous rainout) as well as rainout and snowout. Depending on wind protection the efficiency with which snow samples will be collected may vary. Studies at Cincinnati by Nader (15) show that a polyethylene waste-basket having a diameter of 7.75 inches was as effective in retaining dust as similar containers provided with baffles or wetted with water during periods when the rain had evaporated. The dry solids of rain became attached to the container. Since it is the rain which contains the significant radioactive fallout, this, or a similar container lined with a polyethylene bag, would be a simple and satisfactory fallout collector.

Because most of the activity does not appear in runoff it may be presumed that the radioactivity remains on solid surfaces such as grasses, crops and soil where, although not presenting a signifcant external radiation field, it may, at greatly increased levels, present an internal exposure problem via the food chain.

SUMMARY

Fallout radioactivity, largely as precipitation, has been monitored continuously at Cincinnati, Ohio, since the spring of 1953. Rain, cistern and surface waters were tested to determine distribution and characteristics of the radioactivity.

During the two years of continental testing (1953 and 1955), the predominant air contaminants were characterized as young fission products. The deposition of activity from rain was variable and occasionally quite high. The monthly residual fallout (corrected for decay) reached a maximum of about one microcurie per square meter of surface area on June 1, 1953, and again on June 1, 1955.

In 1954 and 1956 when the air contaminants were of longer half-life the maximum monthly residual was about 0.1 $\mu c/M^2$ in 1954 and 0.4 $\mu c/M^2$ in 1956. It is estimated that on July 1, 1957, the residual activity from fallout during 1953, 1954, 1955, and 1956 was 10, 12, 50 and 105 $m\mu c/M^2$, respectively, or a total of 177 $m\mu c/M^2$ of fission products (9 months to 4 years of age). The strontium-90 (yttrium-90) deposition at Cincinnati for the four years is estimated to be 11 $m\mu c/M^2$ or an average concentration in rain of 3 $\mu\mu c/l$.

Depending on the time of contact with water, the age of the contaminants, and other factors, a variable percentage of the total activity (average of 40 to 50 per cent) is dissolved in rain water. About 40 per cent of the total activity settles out from cistern water. The activity of local surface streams indicates that over 90 per cent of the dissolved and total activity in the rain has been removed. Somewhat less than one-half of the total activity in surface water was in a dissolved state.

Since so little of the rainout activity can be accounted for as runoff it may be presumed that the radioactive fallout is fairly firmly fixed near its point of depositon as particles or by adsorption on solid surfaces such as vegetation and soil.

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